

On the initial stage of quasiparticle decay

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The initial stages of the quasiparticle decay in a Fermi liquid are governed by a time-scale distinct from the scattering rates as derived from the Fermi golden rule approach. We show that the initial decay is non-exponential and that it is determined by the zeroth spectral moment of the electron self-energy. We analyzed numerically a number of approximations for the self-energy by comparing with exact configuration interaction calculations for small finite system with fragmented states. A numerically simple approach for computing the spreading of the quasiparticle states for large systems is devised.

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With the recent spectacular advances in light sources and ultrafast spectroscopic methods it has become possible to trace the quantum dynamics of electronic systems down to the atto-second time scale ([1–3] and references therein). One of the prime goals is the understanding of the nature of the formation and decay of electronic states. In this respect, numerical methods are feasible for few-electron systems only. For condensed matter one has to resort to different concepts such as the Landau's theory of Fermi liquids [4] that describes (low-energy) long-lived excitations as *quasiparticles* (QP). Due to residual interactions QPs decay in time generally. How this decay proceeds in time t is known under certain conditions only: A QP decays exponentially as $\sim \exp(-\gamma t)$ with the rate constant $\gamma(\epsilon) \sim \epsilon^2/\epsilon_F$ [5]. Here ϵ is the quasiparticle energy and ϵ_F is the Fermi energy. This, however, holds true only for times $t \gg 1/\gamma$. The importance of this restriction is illustrated by the following: Let us assume an exponential decay at all t , thus the QP peak appears in frequency with a Lorentzian shape. This means that at the QP energy ϵ the spectral function behaves as $A_L(\omega; \epsilon) = \frac{1}{\pi} \frac{\gamma}{(\omega - \epsilon)^2 + \gamma^2}$. The standard deviation $\sigma^2(\epsilon)$ of the spectral density given by such a functional form diverges,

$$\sigma^2(\epsilon) = \int_{-\infty}^{\infty} d\omega (\omega - \epsilon)^2 A_L(\omega; \epsilon) \rightarrow \infty. \quad (1)$$

Explicit calculations for a three-dimensional (3D) homogeneous electron gas (HEG) show that this divergence is spurious [6] and that the zero, first and second spectral moments are indeed finite. The convergence of the integral Eq. (1) is governed by the high-frequency behavior of the spectral function. Thus, the short-time limit of the single particle Green's function (from which A_L derives) is of special interest. In this respect quantum-kinetics indicates a quadratic decay in time [7]. Experimentally, atto-second resolution of electronic states in condensed matter has already called for a careful inspection of this issue [8].

Here we present a spectral function that exhibits the correct

short and long-time behavior, i.e.

$$\frac{d}{dt} A(t; \epsilon) \xrightarrow{t \rightarrow 0} -\sigma^2(\epsilon)t, \quad (2)$$

$$A(t; \epsilon) \xrightarrow{t \rightarrow \infty} e^{-\gamma t}. \quad (3)$$

These equations are exact and can be obtained nonperturbatively from very general physical considerations [9]. An attempt with a similar goal has been undertaken in Ref. [9], the resulting spectral function, however, violates the sum rules and has a shape with the spectral moments finite at any order, at variance with Ref. [6]. The spectral function given in this work fulfill all sum rules and comply with the exact short and long time-limits. The key ingredients are the imaginary part of the on-shell electron self-energy $\gamma(\epsilon) = \text{Im}\Sigma(\omega = \epsilon; \epsilon)$ and the decay constant $\sigma^2(\epsilon)$ as expressed in terms of the zeroth spectral moment of the self-energy $\Sigma^{(0)} = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega |\text{Im}\Sigma(\omega; \epsilon)|$. The approach provides a recipe to compute the short-time limit of the electron correlation function on the basis of many-particle perturbation theory. In particular, we demonstrate how the decay constant (Eq. 2) can be computed diagrammatically to any desired order in the interaction. Conceptually, the problem should be addressed by the quantum kinetic theory. However in this formalism, analytic calculations of the initial stage of the quasiparticle are not available and numerical approaches rely on further approximations [10–12].

For the decaying part of the spectral function we make an ansatz

$$A(t; \epsilon) = \exp\left(-\gamma(\epsilon) \frac{t^2}{t + \tau(\epsilon)}\right) \quad (4)$$

which obeys the two limiting cases [Eqs. (2,3)] with $\sigma^2(\epsilon) = 2\gamma/\tau$. The analytic form of the Fourier transform of this function ($A(\omega; \epsilon) = \frac{1}{\pi} \int_0^{\infty} \cos[(\omega - \epsilon)t] A(t; \epsilon) dt$) is not known, however it is possible to prove that in the frequency domain it has exactly three finite spectral moments in accordance

with [6]. The large ω expansion reads

$$A(\omega; \epsilon) \sim \frac{6}{\pi} \frac{\gamma}{(\omega - \epsilon)^4 \tau^2} \quad \text{as } \omega \rightarrow \infty.$$

$A(\omega; \epsilon)$ is normalized, i.e., 0th spectral moment is one [13].

According to Altshuler *et al.* [14] the initial stage of the quasiparticle decay always involves a formation of the two-particle-one-hole state ($2p1h$). The rate of the process is given by the *first collision time* $1/\tau_1$ and is determined by the corresponding Coulomb matrix elements or, in other words, by the available phase-space (the energy and the momentum must be conserved). The phase-space also determines in a crucial way the subsequent stages of the QP decay, which results in the creation of an increasing number of particles and holes, forming either localized or delocalized states in a Fock space. In the latter scenario the exponential decay is established after many generations of particles and holes have emerged. From these very general arguments it is obvious that the exponential decay requires a certain time to develop, which in our theory is determined by the parameter $\tau(\epsilon)$. This time parameter certainly exceeds the first collision time ($\tau(\epsilon) \gg \tau_1$) obtained from the golden rule arguments applied to *the bare Coulomb interaction* (at the initial stages the screening is not efficient). This indicates that the time $\tau(\epsilon)$ cannot be obtained from either the bare nor the screened interaction and is distinct from the relaxation time at the large-time limit ($1/\gamma(\epsilon)$).

To obtain $\tau(\epsilon)$ let us recall the relations between the n^{th} order spectral moments $M^{(n)}$ of the single-particle Green function and that of the self-energy $\Sigma^{(n)}$ [6]:

$$M^{(0)} = I, \quad \Sigma_\infty = M^{(1)} - \epsilon, \quad (5)$$

$$\Sigma^{(0)} = M^{(2)} - [M^{(1)}]^2. \quad (6)$$

Σ_∞ is the frequency independent real part of the self-energy [15]. These matrix relations directly follow from the Dyson equation, and can be obtained in any basis ([6] used a plane-waves basis). ϵ is a diagonal matrix with the elements given by the zeroth-order state energies. For finite systems Hartree-Fock basis states are appropriate. Writing the matrix of the spectral functions in terms of the imaginary part of the single-particle Green function ($A(\omega) = \frac{1}{\pi} |\text{Im} G(\omega)|$) and likewise for the spectral function of the self-energy ($S(\omega) = \frac{1}{\pi} |\text{Im} \Sigma(\omega)|$), and using the superconvergence theorem [16] the matrices are cast as frequency integrals:

$$M^{(n)} = \int_{-\infty}^{\infty} d\omega \omega^n A(\omega), \quad n = 0 \dots 2, \quad (7)$$

$$\Sigma^{(0)} = \int_{-\infty}^{\infty} d\omega S(\omega). \quad (8)$$

In HF basis Σ_∞ is rather small and is proportional to the difference of the direct and the exchange Coulomb energy computed with the Hartree-Fock and exact density matrix, i. e. related to the deviation of the natural occupations from 1 or 0. Thus, by virtue of Eqs. (5,7) we arrive at the conclusion that the Hartree-Fock energies in the first approximation are

given by the center of mass of the spectral function. Likewise, by using Eqs. (6,7) we establish a formula for the matrix of standard deviations [cf. Eq. (1)]:

$$\sigma^2 = \int_{-\infty}^{\infty} d\omega S(\omega). \quad (9)$$

Formally an exact representation of this positively defined matrix can be written in terms of the six-point response function [17] (Fig. 1a). Instead, we will compute the standard deviations for finite systems using its factorizations.

To determine the set-in time of the exponential decay for 3D HEG we apply Eq. (9) in a plane-wave basis to the states close to the Fermi surface. Calculations of Vogt *et al.* [6] and Farid [18] show that $\sigma^2(\epsilon)$ has two contributions: i) A local, momentum independent and ii) A non-local, momentum dependent. Their sum remains finite and positive at the Fermi momentum ($\sigma^2(\epsilon_F) > 0$). This tells us that the set-in time for the exponential decay for quasiparticles in the vicinity of the Fermi surface behaves as

$$\tau(\epsilon) = \frac{2\gamma(\epsilon)}{\sigma^2(\epsilon)} \overset{\epsilon \rightarrow \epsilon_F}{\sim} \frac{\epsilon^2}{\epsilon_F \sigma^2(\epsilon_F)}. \quad (10)$$

The prefactor in front of ϵ^2 can be obtained analytically [19]. We also note that for HEG the spectral function consist of a quasiparticle peak with the oscillator strength less the unity (coherent part) surrounded by the satellites (incoherent part) [20]. Our approach goes beyond a description of the quasiparticle peak: fine details of both parts are smeared out preserving, however, the particle number and having a correct asymptotic behavior. Calculation of higher-order satellites for comparison requires inclusion of diagrams of higher-orders in the screened interaction W and is extremely computationally demanding ([21, 22]). Therefore, we verify the performance of different approximations for the self-energy by comparing its zeroth moments computed by the configuration interaction (CI) approach for a *finite electron system*.

As a prototypical system we consider the widely studied Na_9^+ cluster [23–25]. For the current purpose it is advantageous for several reasons: i) it contains a small number of electrons making it accessible to full CI [26, 27], ii) it can be seen as a generalization of 3D HEG to a finite number of particles (jellium model [28]). The σ^2 matrix can be computed exactly by exact diagonalization of the many-body Hamiltonian. We use an algorithm by Olsen *et al.* [29] based on the graphical unitary group approach [30] for the generation of the restricted active space (RAS) and full CI Hamiltonians. The calculations are performed for each spin multiplicity separately using spin-adapted basis functions [31]. In terms of the matrix elements of the creation and annihilation operators the spectral moments are expressed as

$$M^{(n)} = \sum_p^{D^{N+1}} (\epsilon_p^+)^n \mathbf{X}^p \overline{\mathbf{X}}^p + \sum_q^{D^{N-1}} (\epsilon_q^-)^n \mathbf{Y}^q \overline{\mathbf{Y}}^q, \quad (11)$$

where the summation is performed over the Hilbert space of the ionized states (dimension D^{N-1}) and electron attached

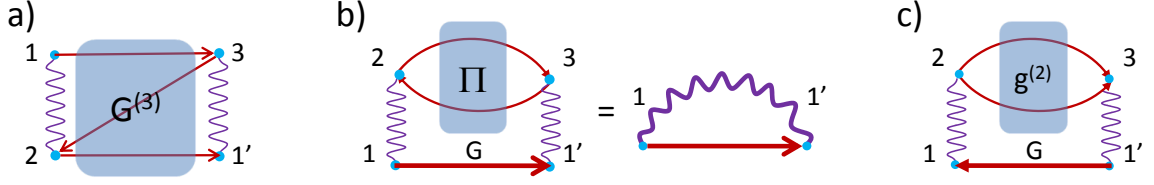


FIG. 1. Diagrams for the electron self-energy. a) exact expression in terms of the $2p1h$ response function in which the entrance and exit channels cannot be separated by cutting one fermion line; b) $(p-h)-p$ factorization leading to the GW approximation; c) $(p-p)-h$ factorization.

states (dimension D^{N+1}). The matrix elements of electron creation (\hat{a}_α^\dagger) and annihilation (\hat{a}_α) operators

$$X_\alpha^p = \langle pN+1 | \hat{a}_\alpha^\dagger | 0N \rangle, \quad Y_\alpha^q = \langle qN-1 | \hat{a}_\alpha | 0N \rangle, \quad (12)$$

and the transition energies $\varepsilon_p^+ = E_p^{N+1} - E_0^N$, $\varepsilon_q^- = E_0^N - E_q^{N-1}$ are computed from the CI many-body states.

Approximations for the self-energy operator can be obtained from the factorization of the $2p1h$ six-points function [15, 17] (Fig. 1a). If the particle-hole $(p-h)$ Green's function is treated exactly we obtain the so-called GW approximation [32] (Fig. 1b). Alternatively, this approximation can be obtained from the $\Psi[G, W]$ variational energy functional [33] expanded in terms of the dressed electron propagator G and the screened Coulomb interaction W . A single diagram of the first order has to be considered. Finally, one obtains the same functional form by neglecting the three-point vertex function Γ in Hedin's equations [34]. It should be noted that we do not perform the self-consistent solution of Hedin's equations; instead, we compute exactly G and W from the exact one-particle and particle-hole propagators. They are given by the Lehmann representations in terms of many-body electron states [35, 36]. In accordance with the spectral representation of the self-energy [26] we obtain for the energy-uncertainty:

$$\Sigma_{\alpha,\beta}^{(0),GW} = 4 \sum_{n \neq 0} \sum_p^{D^N, D^{N+1}} \langle \alpha p | n \rangle \langle n | p \beta \rangle + (p-h)-h \text{ terms}, \quad (13)$$

where we introduced a notation for the convolution of the Coulomb matrix elements $\langle \alpha\beta | \gamma\delta \rangle = \int d(\mathbf{r}_1 \mathbf{r}_2) \phi_\alpha(\mathbf{r}_1) \phi_\beta(\mathbf{r}_1) \phi_\gamma(\mathbf{r}_2) \phi_\delta(\mathbf{r}_2) / |\mathbf{r}_1 - \mathbf{r}_2|$ with matrix elements of the creation (or annihilation) operators [Eq. (12)] and with the density matrix elements $Q_{\gamma\delta}^n = \langle nN | \hat{a}_\gamma^\dagger \hat{a}_\delta | 0N \rangle$:

$$\langle \alpha p | n \rangle = \sum_\beta \sum_{\gamma\delta} X_\beta^p \langle \alpha\beta | \gamma\delta \rangle Q_{\gamma\delta}^n. \quad (14)$$

Analogically, one obtains an expression for the self-energy using the $(p-p)-h$ factorization (Fig. 1 c):

$$\Sigma_{\alpha,\beta}^{(0),G^{(2)}} = \sum_m^{D^{N+2}} \sum_q^{D^{N-1}} \langle \alpha q | m \rangle \langle m | q \beta \rangle + (h-h)-p \text{ terms}, \quad (15)$$

where similar to Eq. (14) we define the convolution of $\langle \alpha\beta | \gamma\delta \rangle$ with the matrix elements of the two creation (annihilation) operators $P_{\gamma\delta}^m = \langle mN+2 | \hat{a}_\gamma^\dagger \hat{a}_\delta^\dagger | 0N \rangle$:

$$\langle \alpha q | m \rangle = \sum_\beta \sum_{\gamma\delta} Y_\beta^q \langle \alpha\beta | \gamma\delta \rangle P_{\gamma\delta}^m. \quad (16)$$

Eqs. (13,15) can be thought of as *the Fermi golden rule* expressions. Since the delta-function ensuring the energy conservation is not present here the whole expression has a dimension of the energy squared. Starting from Eqs. (13,15) we further derive a series of simpler approximations. When the HF Green's function is used in place of G we obtain the so-called G^0W approximation. If, furthermore, the non-interacting excited states are used to compute $Q_{\gamma\delta}^n$ we obtain the G^0W^0 approximation with the spectral moment:

$$\Sigma_{\alpha,\beta}^{(0),G^0W^0} = 2 \sum_{\eta,\gamma,\delta} \langle \alpha\eta | \gamma\delta \rangle \langle \gamma\delta | \eta\beta \rangle n_\gamma (1 - n_\delta), \quad (17)$$

where n_γ is the occupation of the single-particle state γ .

For the Na_9^+ cluster we computed the spectral moments (Eqs. 11,13,15) by exact diagonalization of the many-body Hamiltonian. The exact spectral function (solid line, Fig. 2a) of the lowest valence state ($\varepsilon_1^{\text{HF}} = 9.786$ eV) is fragmented (two major peaks) and has multiple satellites. Despite this fact the model spectral function centered at the HF energy approaches the exact one in a large range of energies. The exact energy-uncertainty from the first two spectral moments of the spectral function (Eq. 11) is compared with the expressions resulting from the approximation for the self-energy (Eqs. 17,13,15) (Fig. 3). Corresponding self-energy spectral functions and weights are in Fig. 2(b,c). Generally, GW self energy yields results superior to other approximations. Despite the fact that $(p-p)-h$ factorization performs bad for $S(\omega)$ (large energy gap in the case of finite systems) the spectral moments are close to those of G^0W^0 approximation.

Summarizing, we presented a form of the quasiparticle line-shape that reflects the correct short and long time-limits of the single-particle Green function and, thus, can be used to parametrize evolving in time electronic structure (e. g., attosecond time-resolved photoemission). The spectral function also explicitly enters a description of a number of static processes (e. g. core state x-ray photoemission). Thus, the experimentally observed form of the Fermi edge singularity will be affected by both the finite life-time of the core state $1/\gamma$ as well as by the corresponding set-in time τ [13].

In the case of 3D HEG the spectral function describes both the coherent and incoherent parts. We show that the set-in time $\tau(\epsilon)$ vanishes as ϵ^2 in the vicinity of the Fermi surface. In the case of finite systems the CI method enables us to compute the energy uncertainties using a number of diagrammatic approximations. Our simulations indicate that accurate results

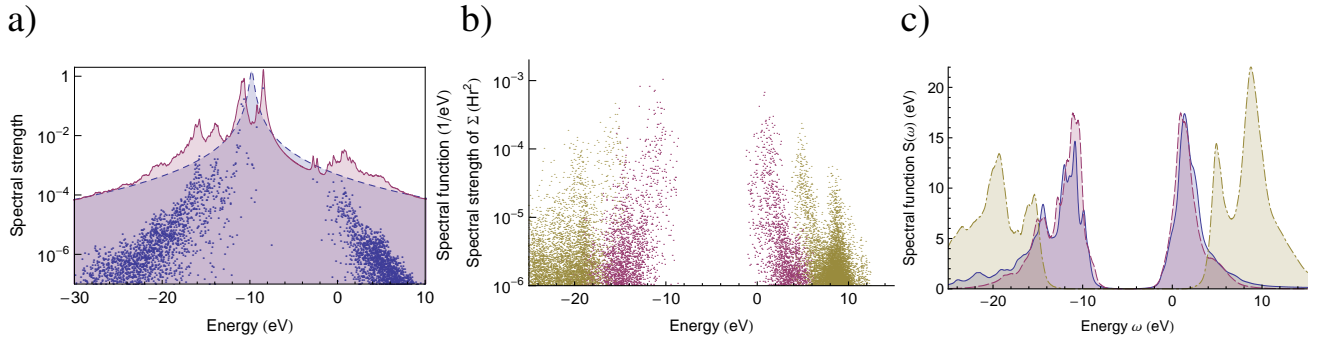


FIG. 2. (Color online) CI calculations for the Na_9^+ cluster: a) The exact (solid) and model (dashed) spectral functions of lowest valence state. The dots denote the weights (squares of the matrix elements, Eq. (12)) of the exact spectral function; b) The spectral strength of the self-energy in $(p-h)-p$ (red) and $(p-p)-h$ (yellow) factorizations; c) The spectral function ($\text{Tr}[\mathbf{S}(\omega)]$) of the exact (solid), $(p-h)-p$ (dashed) and $(p-p)-h$ (dash-dotted) self-energies.

are obtained even by neglecting the three-particle vertex, however, further verifications are needed for extended systems.

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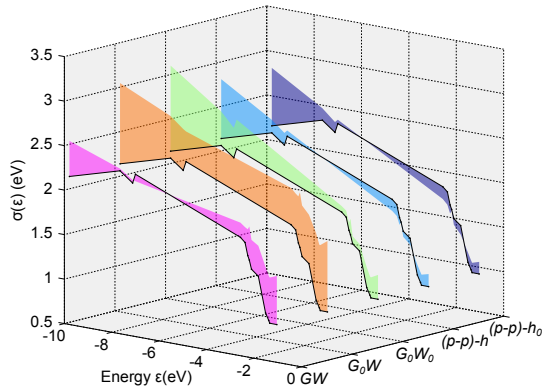


FIG. 3. (Color online) Energy-uncertainty for the Na_9^+ states. Shaded area denotes deviation of the approximate theories from the exact.

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On the initial stage of quasiparticle decay – Supplementary Material –

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General properties

The spectral function of a fermionic many-body quantum system is defined in its most general form as the overlap of a particle (hole) states:

$$2\pi S(t - t'; \epsilon) = \langle \psi(t; \epsilon) | \psi(t'; \epsilon) \rangle.$$

The states emerge by creating a particle for $\epsilon > \epsilon_F$ or a hole with $\epsilon \leq \epsilon_F$ and are not the eigenstates of the system in general and, thus, decay in time. It is convenient to represent the spectral function as a product of a noninteracting oscillatory part and a decaying part $A(t; \epsilon)$:

$$S(t; \epsilon) = \frac{e^{-i\epsilon t}}{2\pi} A(t; \epsilon).$$

For the latter we make an ansatz

$$A(t; \epsilon) = \exp\left(-\gamma(\epsilon) \frac{t^2}{t + \tau(\epsilon)}\right). \quad (1)$$

The Fourier transform of the spectral function can be written as follows:

$$\begin{aligned} A(\omega; \epsilon) &= \int_{-\infty}^{\infty} dt e^{i\omega t} S(t; \epsilon) \\ &= \frac{1}{\pi} \int_0^{\infty} \cos[(\omega - \epsilon)t] A(t; \epsilon) dt. \end{aligned} \quad (2)$$

For $A(t; \epsilon)$ given by Eq. (1) the analytic form of the Fourier transform is not known, but can easily be obtained numerically [Fig. (1)]. Its odd spectral moments are zero because of symmetry consideration $A(t; \epsilon) = A(-t; \epsilon)$:

$$\bar{M}^{(2k-1)}(\epsilon) = \int_{-\infty}^{\infty} d\omega (\omega - \epsilon)^{2k-1} A(\omega; \epsilon) = 0.$$

Thus $\bar{M}^{(1)}(\epsilon) = M^{(1)}(\epsilon) - \epsilon = 0$. The even spectral moments can analytically be obtained from the derivatives of $A(t; \epsilon)$ at $t = 0$:

$$\bar{M}^{(2k)}(\epsilon) = \int_{-\infty}^{\infty} d\omega (\omega - \epsilon)^{2k} A(\omega; \epsilon) = (-1)^k \lim_{t \rightarrow 0} A^{(2k)}(t; \epsilon).$$

This leads to $\bar{M}^{(0)}(\epsilon) = M^{(0)}(\epsilon) = 1$ (normalization condition), and $\bar{M}^{(2)}(\epsilon) = M^{(2)}(\epsilon) - [M^{(1)}(\epsilon)]^2 = \frac{2\gamma(\epsilon)}{\tau(\epsilon)}$. Higher

spectral moments diverge because of the discontinuity of the derivative of $A(t; \epsilon)$ at $t = 0$. This completes the proof that in the frequency domain it has exactly three finite spectral moments in accordance with [1].

The asymptotic large ω expansion can readily be obtained by integrating Eq. (2) by parts :

$$A(\omega; \epsilon) \sim \frac{6}{\pi} \frac{\gamma}{(\omega - \epsilon)^4 \tau^2} \quad \text{as } \omega \rightarrow \infty.$$

Fermi edge singularities

The proposed spectral function can be applied to obtain more accurate descriptions of the x-ray photoemission (XPS) line shapes from core states. The theory describing the many-electron response in XPS originated in works of Anderson [2, 3], Mahan [4], Nozières and DeDominicis [5]. It was shown that a creation of an infinite number of electron-hole pairs accompanies the photoemission event. The result-

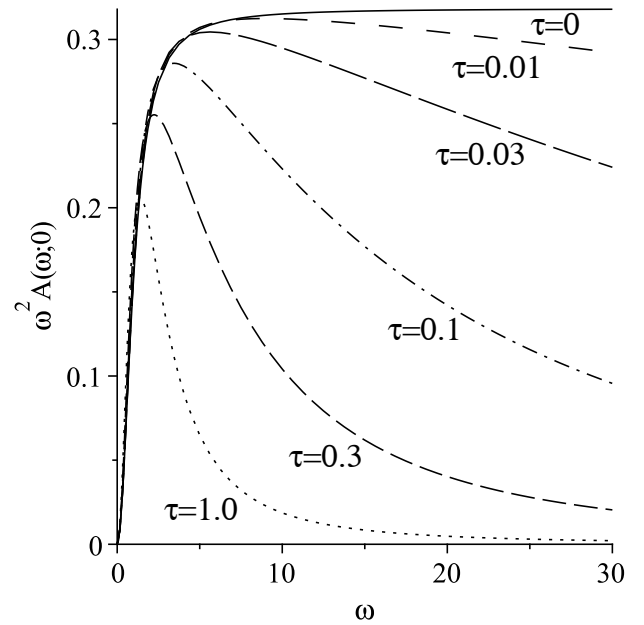


FIG. 1. $\omega^2 A(\omega; 0)$ for the model spectral function defined by Eq. (1) for the rate constant $\gamma = 1.0$ and different values of the set-in time.

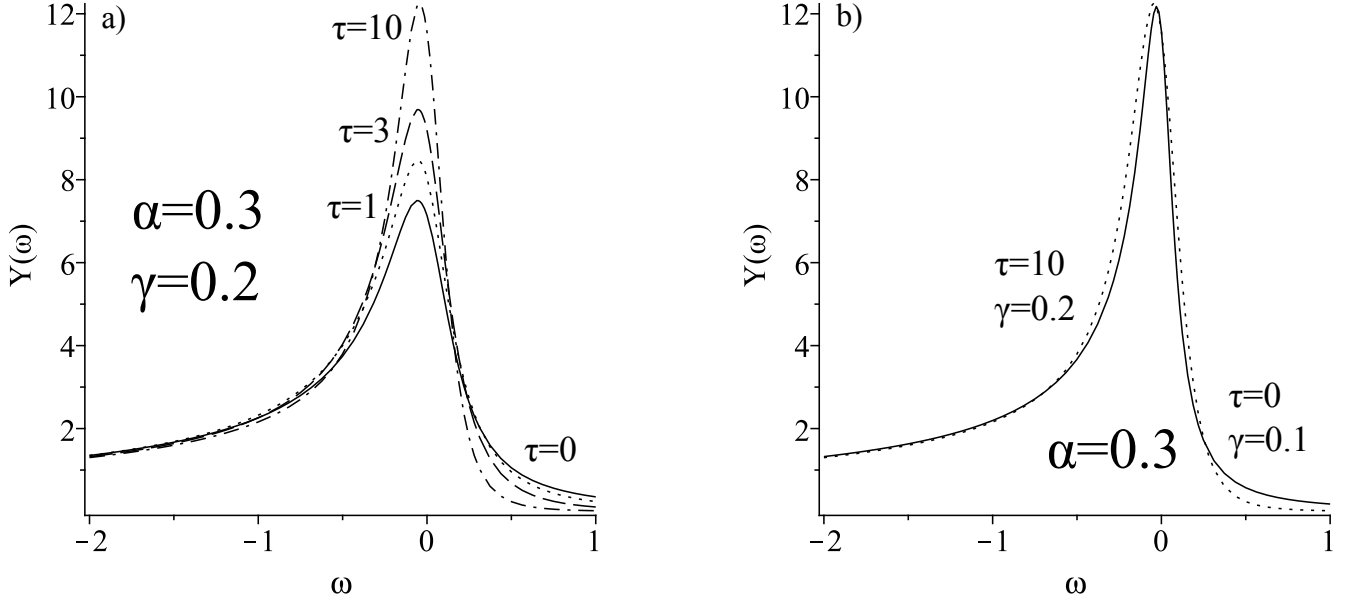


FIG. 2. a) Line-shapes for the x-ray photoemission from core states with finite core state broadenings γ and different set-in times τ . $\tau = 0$ (solid line) corresponds to the Doniach and Šunjić (DS) line-shape. b) The effect of finite set-in time (dotted) can results in a XPS profile experimentally indistinguishable from the DS line-shape with a substantially reduced broadening.

ing electron spectrum for an infinitely-long-lived core hole is given by $\frac{2\pi}{\Gamma(\alpha)} \frac{\theta(-\omega)}{\omega^{1-\alpha}}$, where $0 < \alpha < \frac{1}{2}$ is called the Anderson singularity index and can be given in terms of the scattering phase shifts δ_l as

$$\alpha = 2 \sum_l (2l+1) \left(\frac{\delta_l}{\pi} \right)^2.$$

For brevity we set the origin of the energy scale at the no-loss position of the hole-state.

This approach is oversimplified and a proper treatment requires a finite lifetime of the core holes to be taken into account. Convolution of the singular line-shape with a simple decay function $\exp(-\gamma t)$ yields the well-known Doniach and Šunjić[6] profile:

$$Y^0(\omega) = \frac{2\Gamma(1-\alpha)}{(\omega^2 + \gamma^2)^{\frac{1-\alpha}{2}}} \cos \left[\frac{1}{2}\pi\alpha + (1-\alpha) \arctan \left(\frac{\omega}{\gamma} \right) \right],$$

where Γ is the gamma function.

It is clear that even this modification is insufficient as it is based on the wrong assumption of the hole's spectral function. Therefore, we propose a modified line shape based on the presented spectral function with correct asymptotic behavior.

$$Y(\omega) = \lim_{\delta \rightarrow +0} \int_{-\infty}^{\infty} \exp \left(-\frac{\gamma t^2}{\tau + |t|} \right) e^{i\omega t} \frac{dt}{(\delta - it)^\alpha}, \quad (3)$$

where infinitesimally small positive δ was introduced to select a branch of the multivalued $1/(it)^\alpha$ function and to facilitate the numerical integration (Fig. 2).

A comparison of XPS line-shapes for different set-in times (Fig. 2, b) reveals a similarity between the effect of finite set-in time and that of the reduced broadening. In other words, while γ leads to the broadening of spectral lines the presence of non-zero set-in time τ has an opposite effect as evidenced by $\bar{M}^{(2)}(\epsilon) = \frac{2\gamma(\epsilon)}{\tau(\epsilon)}$. This observation might have a strong impact on the interpretation of the experimental XPS spectra.

We also note a different approach leading to a modified spectral line-shape, the assumption of the frequency dependent scattering phase as was recently implemented for low-dimensional systems by Mkhitarian and Raikh [7].

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